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MAGNETOSTRICTION CONSTANTS

IN IRON-COBALT ALLOYS

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MAGNETOSTRICTION CONSTANTS IN IRON-COBALT ALLOYS*

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ABSTRACT

The single crystal magnetostriction constants in the iron-cobalt system from 0-70% cobalt have been measured using the resistance strain gauge method. Small single crystals in the form of oblate spheroids were employed for the measurements. In the composition range for which no single crystals could be obtained, the constants were obtained from polycrystalline measurements using the method of Bates and Lee. The results obtained by the two methods give continuous curves for the two significant magnetostriction constants λ_{100} and λ_{111} over the entire composition range of interest. The curve for λ_{100} reaches a maximum near 30% cobalt and gives a value of 150×10^{-6} which represents the highest measured magnetostriction constant obtained in a metallic sample; and the curve for λ_{111} goes through zero at approximately 40% cobalt which is near the composition at which the magnetic crystalline anisotropy reverses sign.

INTRODUCTION

High permeabilities at low and high inductions can be obtained in

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iron-cobalt alloys in the composition range of 35 to 50% cobalt. In this composition range, the sign of the magnetic crystalline anisotropy reverses and the saturation moment is a maximum. The direction of easy magnetization changes at 42% from the [100] on the iron-rich side of this critical composition to the [111] direction on the cobalt-rich side. In many alloys in which the anisotropy energy thus goes through zero, e.g., iron-nickel, iron-silicon, the magnetostriction goes through zero at some nearby composition. This has often been interpreted as suggesting a relationship between the anisotropy and magnetostriction and also providing a basis for a theory of the relatively high permeabilities found in these composition ranges.(1) In iron-cobalt alloys, however, the only available magnetostriction data has been obtained in polycrystalline material and the values at saturation are large over the entire composition range and do not suggest any unusual behavior of the magnetostriction. The quantity of interest in the theory of permeability and coercive force(2) as well as in some of the theories of the behavior of Alnico V(3) and the effect of ordering on magnetic properties(4) is the distortion of the unit cell under magnetization in various directions. It appeared desirable, therefore, to measure the single crystal magnetostriction constants in a series of iron-cobalt alloys of varying composition.

An attempt to grow single crystals by the strain anneal method proved fruitless. We were fortunate, however, in having made available to us, through the courtesy of Professor L. W. McKeehan, the single crystals used by Shih in the original investigation(5) of the magneto-crystalline anisotropy

constants in the iron-cobalt system. Although the crystals were quite small in over-all dimensions, the development of the strain gauge technique for the measurement of magnetostriction and the availability of extremely fine gauges has made possible measurements on very small crystals to a relatively high degree of accuracy.

An alternative method of obtaining the single crystal constants from measurements on the polycrystalline material has recently been suggested by Bates and Lee.[6] They find for the case of well-annealed nickel that if the polycrystalline magnetostriction is plotted as a function of the quantity $\frac{M}{M_s}\left(\frac{2M}{M_s} - 1\right)$, the slopes of the resulting curves give λ_{100} and λ_{111} respectively. Although there is no a priori theoretical reason for the validity of this method, it appears to have given satisfactory agreement with data obtained from single crystal measurements in the case to which the method has been applied. In order to extend the results of our investigation to include composition ranges in which no single crystals were available or could be obtained, we investigated the applicability of the Bates-Lee method to our alloys. To this end, measurements of the polycrystalline magnetostriction as a function of induction were made at one composition in the range where single crystal data were available and at some of the other compositions where they were not. The good agreement obtained by the two methods in the former case lends support to the validity of the method and, together with the data at other compositions, has made it possible for us to obtain a continuous

curve for the two significant magnetostrictive constants over the entire range of stability of the α-phase of the iron-cobalt system, i.e., from 0 to 70 percent cobalt.

EXPERIMENTAL PROCEDURE

1. Measurements on Single Crystals

The resulting strain in a single crystal of a ferromagnetic material that has been placed in a magnetic field sufficient for saturation can be expressed as a power series in the angles made by the strain and field direction with the crystal axes. For a crystal of cubic symmetry, Becker has represented the distortion by the following relation:(7)

$$\begin{aligned}\frac{dl}{l} = {} & h_1\left(\alpha_1^2\beta_1^2 + \alpha_2^2\beta_2^2 + \alpha_3^2\beta_3^2 - \tfrac{1}{3}\right) + h_2\left(2\alpha_1\alpha_2\beta_1\beta_2 + 2\alpha_1\alpha_3\beta_1\beta_3 + 2\alpha_2\alpha_3\beta_2\beta_3\right) \\ & + h_4\left(\alpha_1^4\beta_1^2 + \alpha_2^4\beta_2^2 + \alpha_3^4\beta_3^2 + \tfrac{2}{3}\left[\alpha_1^2\alpha_2^2 + \alpha_1^2\alpha_3^2 + \alpha_2^2\alpha_3^2\right] - \tfrac{1}{3}\right) \qquad (1) \\ & + h_5\left(2\alpha_1\alpha_2\alpha_3^2\beta_1\beta_2 + 2\alpha_1\alpha_2^2\alpha_3\beta_1\beta_3 + 2\alpha_1^2\alpha_2\alpha_3\beta_2\beta_3\right) + \tfrac{1}{3}\left.\frac{\Delta V}{V}\right|_{\alpha_i}\end{aligned}$$

where the α_i and β_i are the direction cosines (with respect to the crystal axes) of the direction of strain measurement and of the magnetic field respectively. This equation also gives the magnetostriction of each domain (excluding boundaries) when the crystal is in zero external field; but then the resultant strain of the whole crystal depends on the directional distribution of the individual domain magnetization vectors. In order to avoid this uncertainty in specifying the magnetostriction for the demagnetized state, differences in strain for various directions of the saturation field can be used to evaluate the constants in the equation. When the volume

term and the constants h_4 and h_5 are small, the strains in two given lattice directions need be measured for only two field directions each in order to evaluate h_1 and h_2.

In this experiment the magnetostriction was measured with the strain gauge technique.(8) Type A-19 strain gauges, made of fine advance wire bonded in paper, were cemented to the sample in a given crystallographic direction with Duco cement. Since the sections of gauge wire made angles of about 7° with one another (see fig. 1), the direction midway between them was used as the gauge direction. Errors due to the resulting angular uncertainty are discussed below.

The crystal orientation with respect to a fiducial mark on its surface was determined by means of Laue back-reflection X-ray patterns; and the gauge was readily affixed with respect to the fiducial mark with the aid of a low-power microscope having a vernier-controlled rotating cross-hair. The single crystal specimens of 40, 50 and 70% cobalt in iron were small oblate spheroids with a (110) equatorial plane, and with dimensions approximately a = b = 3mm.; c = 0.2mm. as indicated in figure 1. The X-ray patterns of these crystals indicated pronounced mosaic structure so that the crystals cannot be considered completely perfect. In fact, further careful X-ray examination of a fourth crystal containing 30% cobalt showed it to be a bi-crystal so that the constants for this crystal, previously reported by one of us(9), are probably in error.

The strain gauge on the sample was the fourth arm of a Wheatstone

bridge, the other three arms being similar gauges cemented to a block of constantan. The whole, enclosed in a suitable brass and plastic container, was placed in the magnetic field, thereby minimizing effects due to magnetoresistance and thermal fluctuations. (A diagram of the sample and enclosure is shown in Fig. 2) The output from the bridge was amplified by a 75 cycle Liston-Folb D.C. chopper amplifier, and the amplifier output fed to an Esterline-Angus recording milliameter. The bridge was balanced and the output calibrated by means of a standard resistance box in parallel with the sample gauge. These particular A-19 gauges had a resistance of 60 ± 0.5 ohms, and a gauge factor of $1.62 \pm 3\%$ where the gauge factor is defined as the ratio of $\frac{\Delta R}{R}$ to the strain $\frac{\Delta l}{l}$.

Carr[10] has calculated the error in measurement due to uncertainty in the relative orientations of the crystal axes, the magnetic field, and the strain gauges. He finds for a (110) plane in which the final magnetization direction makes the angle θ and the direction of strain measurement makes the very small angle $\Delta\varphi$ with the initial magnetization direction, and for an initial magnetization which lies very close to the [001] direction, therefore having direction cosines n_1, n_2 and 1:

$$\left.\frac{dl}{l}\right|_{initial} - \left.\frac{dl}{l}\right|_{final} = h_1 \sin^2\theta - \frac{1}{\sqrt{2}}(\Delta n_1 - \Delta n_2)(h_1 - h_2)\sin 2\theta - \Delta\varphi h_2 \sin 2\theta \qquad (2)$$

Then the error is minimum for $\theta = 90^\circ$; i.e., the difference in strain between the two field settings parallel and perpendicular to an [001] directed gauge in the (110) plane is a direct measure of the constant $h_1 = \frac{3}{2}\lambda_{100}$. In a similar manner the best value of $h_2 = \frac{3}{2}\lambda_{111}$ can be obtained in the (110) plane with a [$\bar{1}$11] directed gauge, when the initial and final field settings

make 45° and 135° respectively with the [001] direction. Then

$$\left.\frac{dl}{l}\right|_{initial} - \left.\frac{dl}{l}\right|_{final} = \frac{2}{3} h_2 (\sqrt{2} - \Delta\varphi) \simeq \frac{2}{3}\sqrt{2}\, h_2 . \qquad (3)$$

In the measurement of h_1 the quantity $5h_4/6$ is neglected and in the measurement of h_2 the quantity $\frac{h_5}{4}$ is neglected. However, the error just discussed, for angles other than those chosen above, was found sufficient to mask effects due to h_4 and h_5; and for this reason h_4 and h_5 are assumed small. The form effect can be neglected because of the favorable dimensions ratio.

2. Measurements on Polycrystalline Materials

The magnetostriction of polycrystalline iron-cobalt alloys has been investigated by S. R. Williams[11] and by Y. Masiyama[12] using ellipsoidal or rod-shaped specimens. However, there may have been considerable non-randomness in the domain and crystallite orientations of these samples in the demagnetized state even though they were well-annealed, leading to unknown uncertainties in the values of the measured magnetostriction. To eliminate these effects, we have repeated these measurements using cubic specimens and have measured the magnetostriction in three orthogonal directions.

The 0.86 inch cubes, cut from 1x1x30 inch forged bars of iron-cobalt alloys, had sides parallel to better than 5×10^{-4} inches. They were annealed in argon for two hours at 1000°C., cooled in the furnace to 40°C. and then cooled in air. Their exact dimensions and compositions are given in Table 1. Masiyama's specimens, whose chief impurities were reported as traces, were annealed at 1050°C. in a vacuum for 15 hours, then slowly cooled. Williams used specimens made by mixing the powdered elements obtained from the reduced

oxides, pressing, sintering and hot swaging followed by an annealing at 1000°C in hydrogen for two hours, and slow cooling. No analyses were given.

The circuit diagram of the magnetostriction measuring apparatus and specimen holder are illustrated in Figure 3. A resistance strain gauge was glued to the cube surface at the midpoint of the center line. Error in the measured magnetostriction due to slight deviation of the gauge direction from the center line is negligible. A coil to measure magnetization was wound about the cube very close to the gauge. The cube was placed in a thick-walled brass box which had two side walls of very thin Hipernik plate in good contact with the cube and with the magnet pole pieces, and then the box was covered with a very thick cork layer. These devices protected the cube from thermal fluctuations in the surrounding atmosphere and allowed good magnetic and thermal contact with the magnet pole pieces. Thus the demagnetizing factor was eliminated, and the large heat capacity of the magnet prevented thermal variation in the sample due to the magnetocaloric effect and Joule heat in the magnetizing coil. The effect of magnetoresistance in the gauges was negligible since the magnetostriction was rather large and the fields not high. Three fixed resistances made of advance wire were kept in a dewar along with the dry cell. The dewar and a variable resistance box used to balance the bridge were packed in glass wool and placed in a box of heavy aluminum plate. The unbalance voltage from the bridge was fed to a photoelectric galvanometer. The induction in the specimen was measured with a coil wound about the sample connected to a ballistic galvanometer of high sensitivity. Both specimen and magnet were carefully demagnetized before each run. Ambient temperature changes were reduced to a minimum during each run.

3. Results

Table 2 gives the values of h_1 and h_2 for the three single crystals of 40, 50 and 70% cobalt obtained by the above method as well as the calculated values of λ_s, the saturation polycrystalline magnetostriction, obtained from the formula

$$\lambda_s = \frac{2\lambda_{100} + 3\lambda_{111}}{5} = \frac{4h_1 + 6h_2}{15} \quad . \qquad (4)$$

Values of λ_s taken from the work of Masiyama and Williams are included for reference. These data are plotted as a function of cobalt content in Fig. 4.

In Fig. 5, the magnetostriction curves of the polycrystalline cubes of 20.3, 32.3 and 60.5% cobalt in iron respectively are plotted against field strength. In each case three curves are given, one for each of the three principal cubic directions. Only in the case of the 20.3% alloy where the magnetostriction at saturation in the third direction differs by about 8% from the other two, is there any evidence for preferred orientation of either domains or crystallites. The resulting average values are given in Table 3, and a comparison with published values also made.

It is to be noted that the curves for 20.3 and 32.3% cobalt have negative slopes near saturation, suggesting negative λ_{111} as predicted by the single crystal data. This region near saturation is the region where the domain magnetizations rotate from an easy direction nearest the field direction into the field direction; in these two cases, having less than 42% cobalt, the rotations proceed from the [100] direction nearest the

field, so that the magnetostriction has a strong dependence on λ_{111}. Above 42% cobalt, [111] is the easy direction, so that near saturation the polycrystalline magnetostriction has a strong dependence on λ_{100}.

Curves for the polycrystalline magnetostriction plotted against $\frac{M}{M_s}\left(\frac{2M}{M_s} - 1\right)$ for cubes of o0.5, 32.3, and 20.3% cobalt are given in Fig. 6. The only region of the magnetization curve where one can say with certainty that only one mechanism of magnetization is operating is that near saturation where the domains are rotating from the direction of easy magnetization to that of the field. Only those crystals with the applied field in the hardest direction are still contributing to the magnetization at inductions very near saturation. Therefore, only that magnetostriction constant associated with magnetization in the hard direction (h_2 in the case of the 20.3 and 32.3 percent alloy and h_1 in the 60.5% alloy) is important.

At lower inductions, there is overlap between the various operating mechanisms and one cannot therefore ascertain unambiguously which constants are contributing to the magnetostriction. The slope at high inductions for the curve for the 60.5% alloy in Fig. 6 gives very good agreement with the single crystal data as noted in Table 2 and Fig. 4. If one combines the constant thus obtained from the high induction data with the measured polycrystalline saturation magnetostriction, the values for the other constant (λ_{100} in the 20.3 and 32.3% alloy and λ_{111} in the 60.5% alloy) are obtained using Equation (4) and these are given in Table 3. These constants together with those obtained from the single crystal data and those quoted by

Becker and Döring for pure iron form two continuous curves over the entire range of composition of the alloy system in which the body-centered phase is the stable one. These curves are given in Figure 4 together with the measured saturation magnetostriction data.

DISCUSSION

The curve for λ_{100} based on the three single crystal measurements is still rising with decreasing cobalt content at the lowest cobalt composition for which single crystal data are available. If one puts in the additional points obtained from the polycrystalline data by the method described one observes that the curve reaches a maximum at approximately 34% cobalt. This is nearly identically the composition at which the saturation magnetization is a maximum. If h_1 between 0 and 45% cobalt is due to simple dipole-dipole interactions, which depend on the square of the magnetic moment, this result would not be completely unexpected. At the maximum, the magnitude of h_1 is $\sim 230 \times 10^{-6}$ which we believe to be the highest value yet obtained in a metal. If a single crystal could be obtained at this composition and the domains given a preferred direction perpendicular to a [100] direction of strain measurement, a value of magnetostriction comparable to h_1 would be measurable. Steps to approximate this situation by suitably orienting the grains of a polycrystalline material have been undertaken in this laboratory. It is interesting to note that the highest previously reported measurement of magnetostriction in a ferromagnetic metal was that of Nesbitt[13] who studied the magnetostriction of a cold-worked tape containing 70% cobalt and reported a value

for $\left(\frac{\Delta l}{l}\right)_{saturation}$ of 135×10^{-6}. Bozorth[14] has interpreted this as arising from the preferred orientation of the domains perpendicular to the rolling direction which would give a magnetostriction of $\frac{3}{2}\lambda_s$. Clearly, this is an oversimplification since the actual magnitude of the observed magnetostriction must depend very significantly on the crystal orientation, domain orientation and the magnitudes of h_1 and h_2, all of which were not well-known to the investigators. It is thus a fortuitous circumstance that these very high values obtained for λ_s and $\left(\frac{\Delta l}{l}\right)_{saturation}$ in the tape are found at 70% cobalt. At the lower cobalt content where we find a maximum in h_1, the negative h_2 tends to reduce the overall saturation magnetostriction whereas at 70% cobalt, although the value of h_1 is considerably smaller, h_2 is of the same order of magnitude and the two combine to give a larger λ_s.

In the case of h_2, the measured single crystal values together with the known data for iron suggest that h_2 will be negative at the iron-rich side of the alloy system. This is confirmed by the polycrystalline measurements. On the basis of the resulting curve embracing all the points, which is plotted in Figure 4, we have placed the cross-over point at 39% cobalt. This is very close to the cross-over point in the anisotropy energy vs. composition which Shih finds to be at 42% cobalt. The theory is not sufficiently advanced to warrant hazarding an explanation, if one exists, for this remarkable circumstance. It might appear that both the anisotropy and magnetostriction similarly originate in spin-orbit interactions. If this agreement is not merely a fortuitous circumstance, it does suggest that a simple dipole theory is insufficient to explain the many facets of the magnetostriction problem.

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TABLE I

	COMPOSITION							DIMENSION
	Co	Fe	Si	Mn	C	P	S	
SPECIMEN I	60.51	bal	tr	0.27	0.014			2.168x2.168x2.163 cm
SPECIMEN II	32.31	bal	tr	0.13	0.011			2.167x2.168x2.172
SPECIMEN III	20.30	bal	0.009	0.017	0.012	0.008	0.009	2.166x2.167x2.167

TABLE II

% Cobalt	h_1	h_2	λ_s calculated	λ_s Masiyama	λ_s Williams
70	$12.2\ 10^{-5}$	$10.5\ 10^{-5}$	$7.5\ 10^{-5}$	$8.9\ 10^{-5}$	$5.7\ 10^{-5}$
50	$17.9\ 10^{-5}$	$6.2\ 10^{-5}$	$7.2\ 10^{-5}$	$6.8\ 10^{-5}$	$6.5\ 10^{-5}$
40	$22.0\ 10^{-5}$	$1.3\ 10^{-5}$	$6.4\ 10^{-5}$	$6.4\ 10^{-5}$	$6.4\ 10^{-5}$

TABLE III

Cobalt Content %	observed	λ_{100}	λ_{111}	h_1	h_2
20.3	33.1×10^{-6} Writer 2400 oer 30.6×10^{-6} Williams 1200 oer 29.0×10^{-6} Masiyama 1100 oer	$\frac{5}{2}(\lambda_s - \frac{3}{5}\lambda_{111}) = +135.0\times10^{-6}$	-34.8×10^{-6} observed in high field	$\frac{3}{2}\lambda_{100} = 202.5\times10^{-6}$	$\frac{3}{2}\lambda_{111} = -52.2\times10^{-6}$
32.3	43.3×10^{-6} Writer 2400 oer 46.7×10^{-6} Williams 1200 oer 46.5×10^{-6} Masiyama 1100 oer	$\frac{5}{2}(\lambda_s - \frac{3}{5}\lambda_{111}) = +153.3\times10^{-6}$	-30.0×10^{-6} observed in high field	$\frac{3}{2}\lambda_{100} = 230.0\times10^{-6}$	$\frac{3}{2}\lambda_{111} = -45.0\times10^{-6}$
60.5	71.6×10^{-6} Writer 2400 oer 67.1×10^{-6} Williams 1200 oer 72.9×10^{-6} Masiyama 1100 oer	90.0×10^{-6} observed in high field	$\frac{5}{3}(\lambda_s - \frac{3}{5}\lambda_{100}) = 59.3\times10^{-6}$	$\frac{3}{2}\lambda_{100} = 135.0\times10^{-6}$	$\frac{3}{2}\lambda_{111} = 89.0\times10^{-6}$

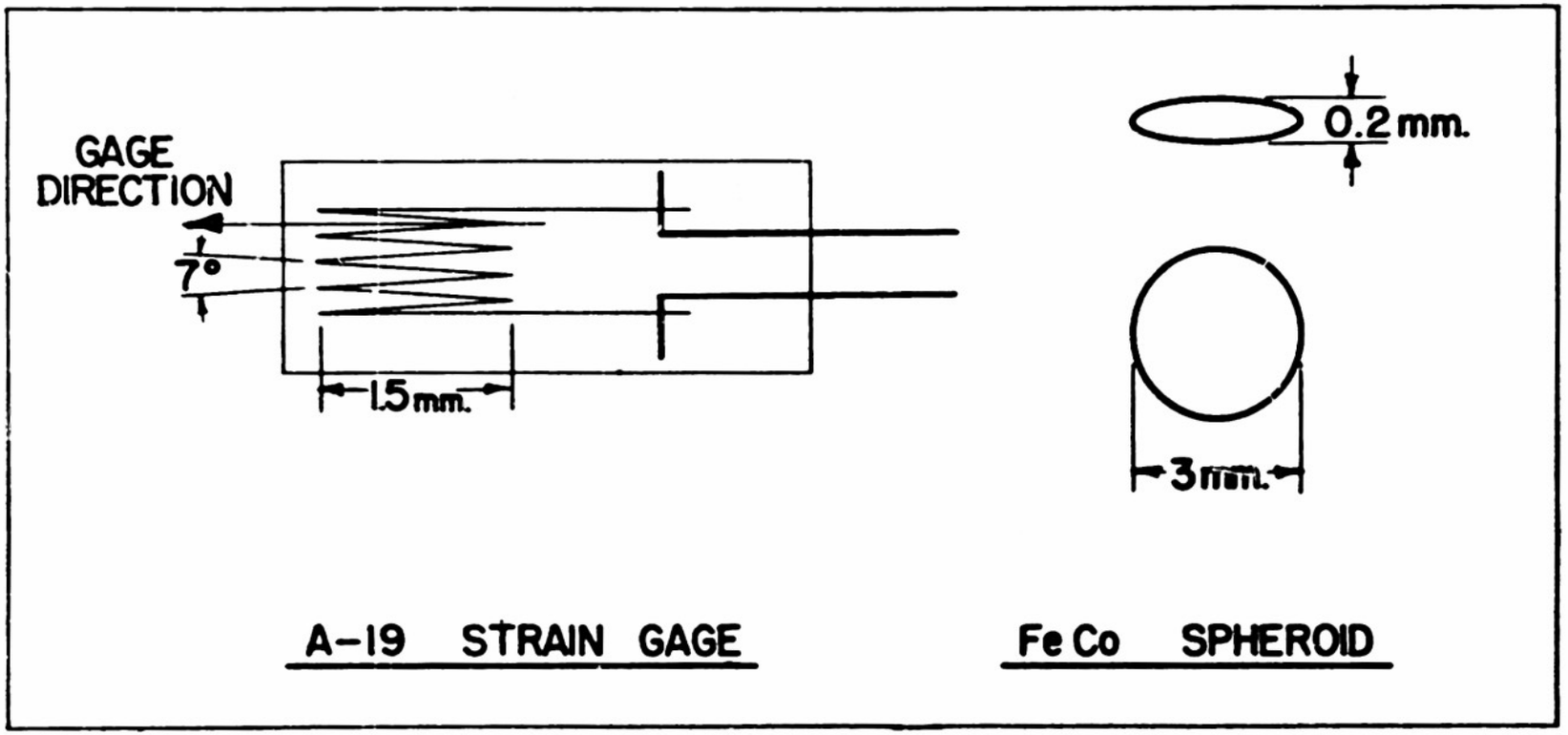

Figure 1. Diagram of an FeCo ellipsoid and of an A-19 strain gage.

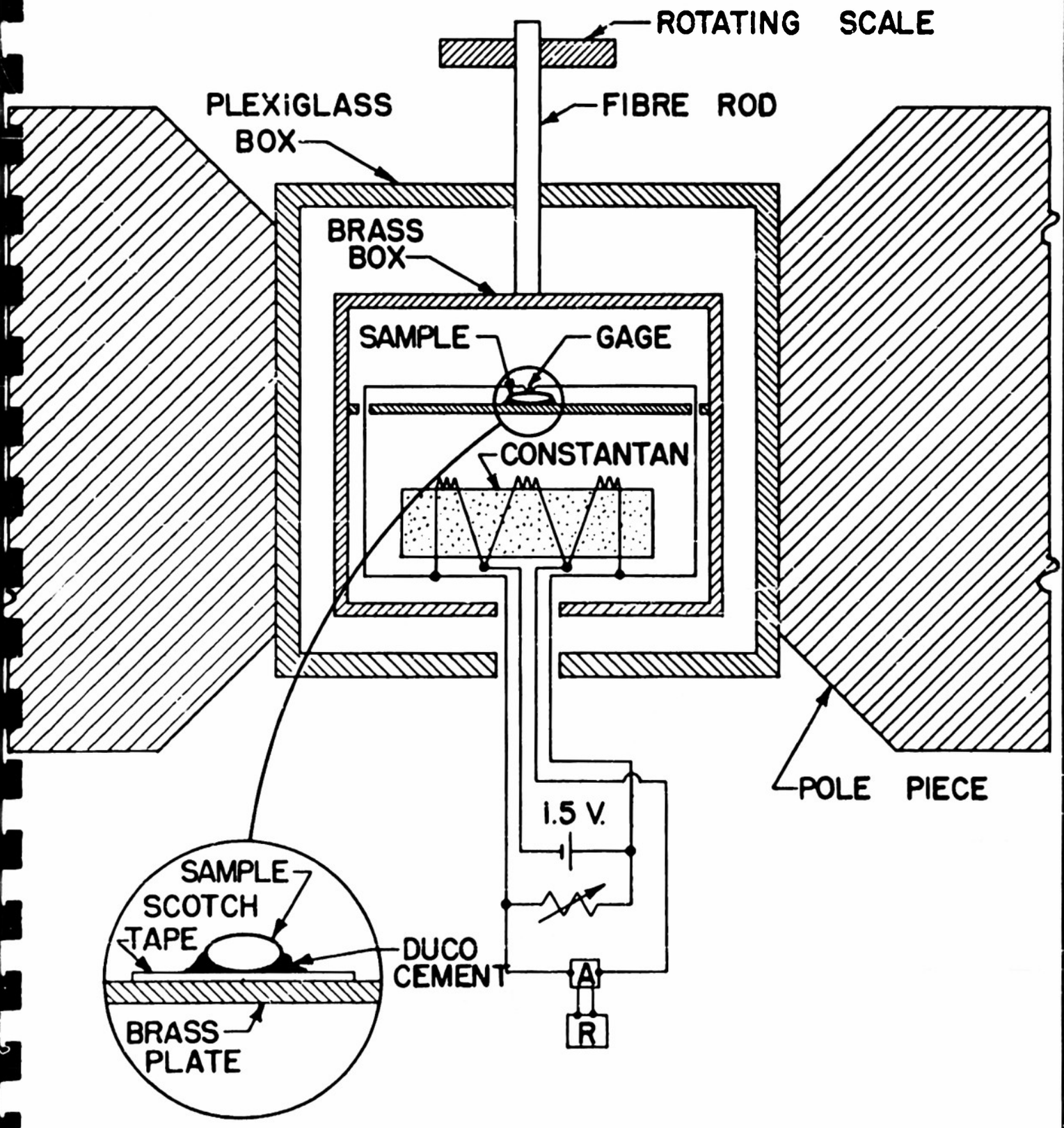

Figure 2. Diagram of the crystal holder and single crystal magnetostriction measuring apparatus.

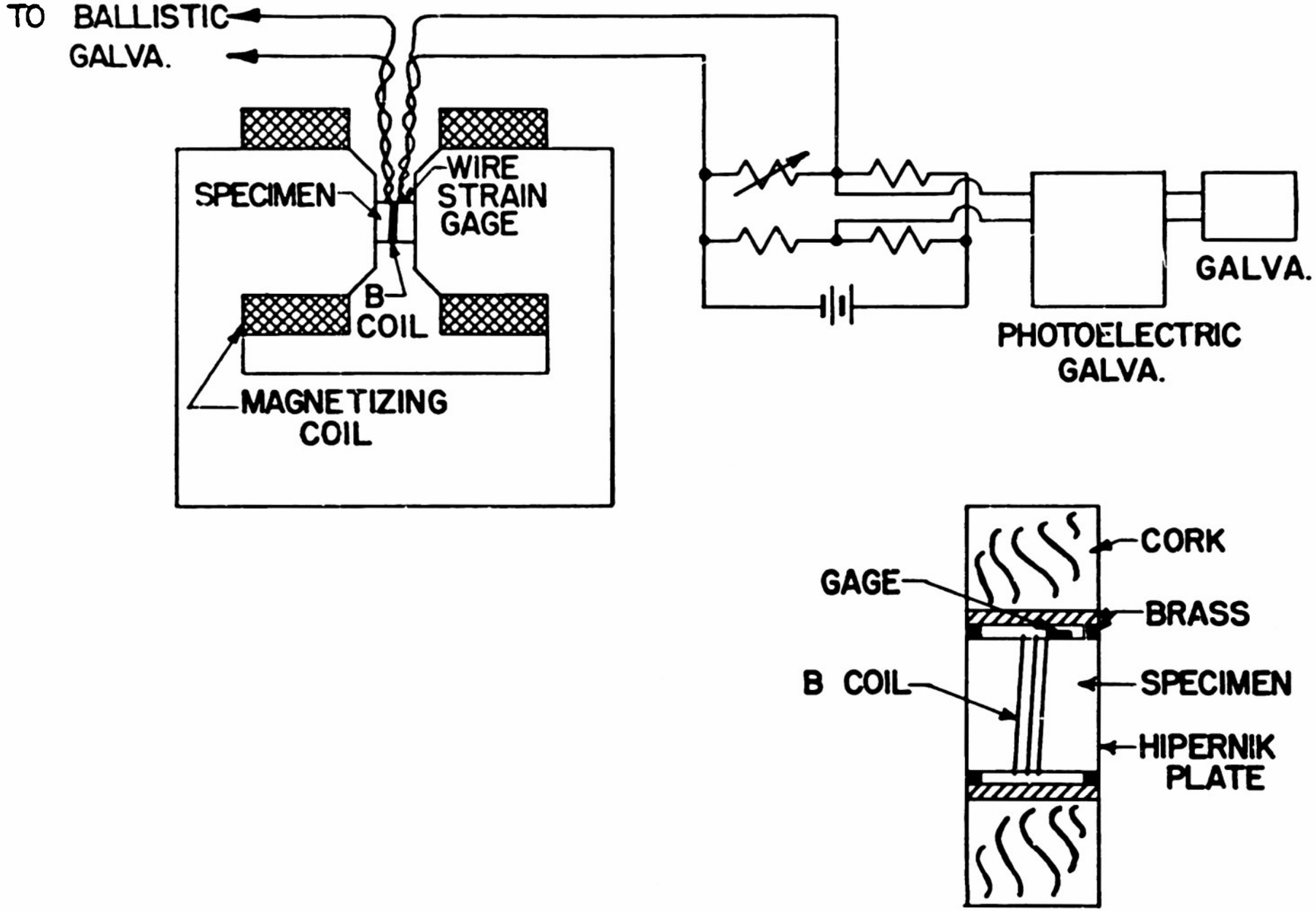

Figure 3. Di_gram of the polycrystalline magnetostriction measuring apparatus and specimen holder.

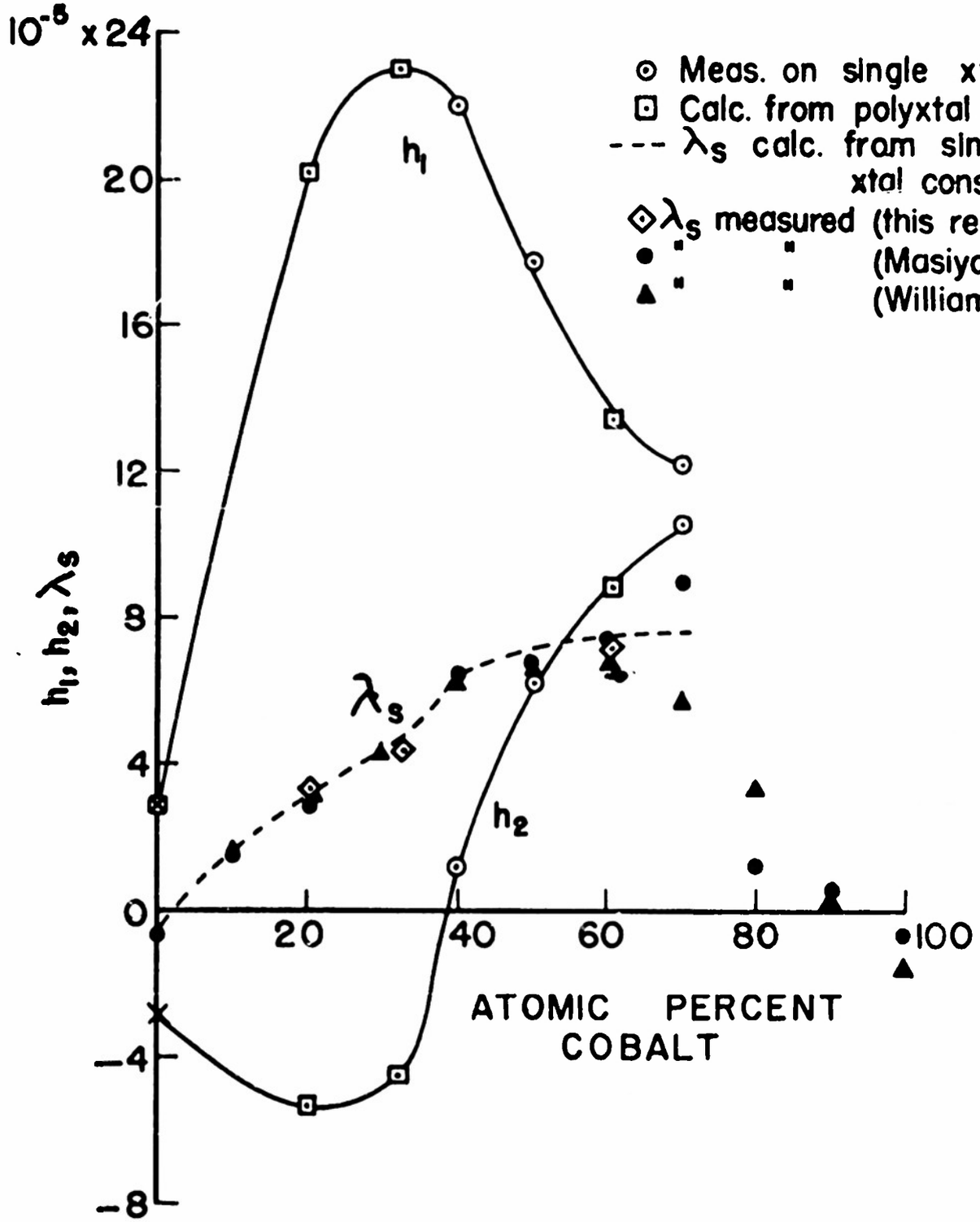

Figure 4. Magnetostriction constants of iron-cobalt alloys from single crystal and polycrystalline data. The points given for pure iron are those selected by Becker and Doring as the most accurate measured values.

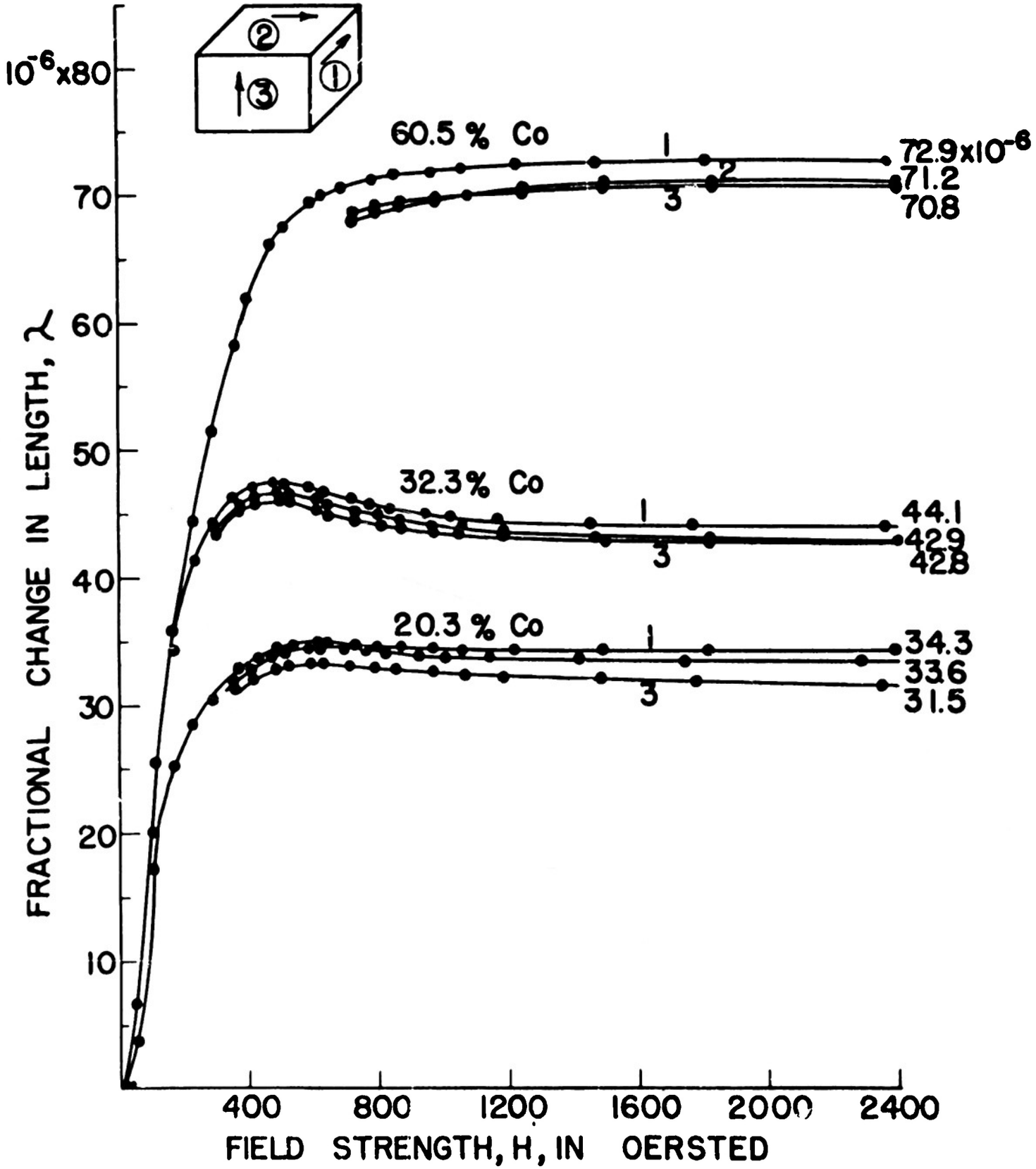

Figure 9. Magnetostriction of polycrystalline iron-cobalt alloys as a function of field intensity.

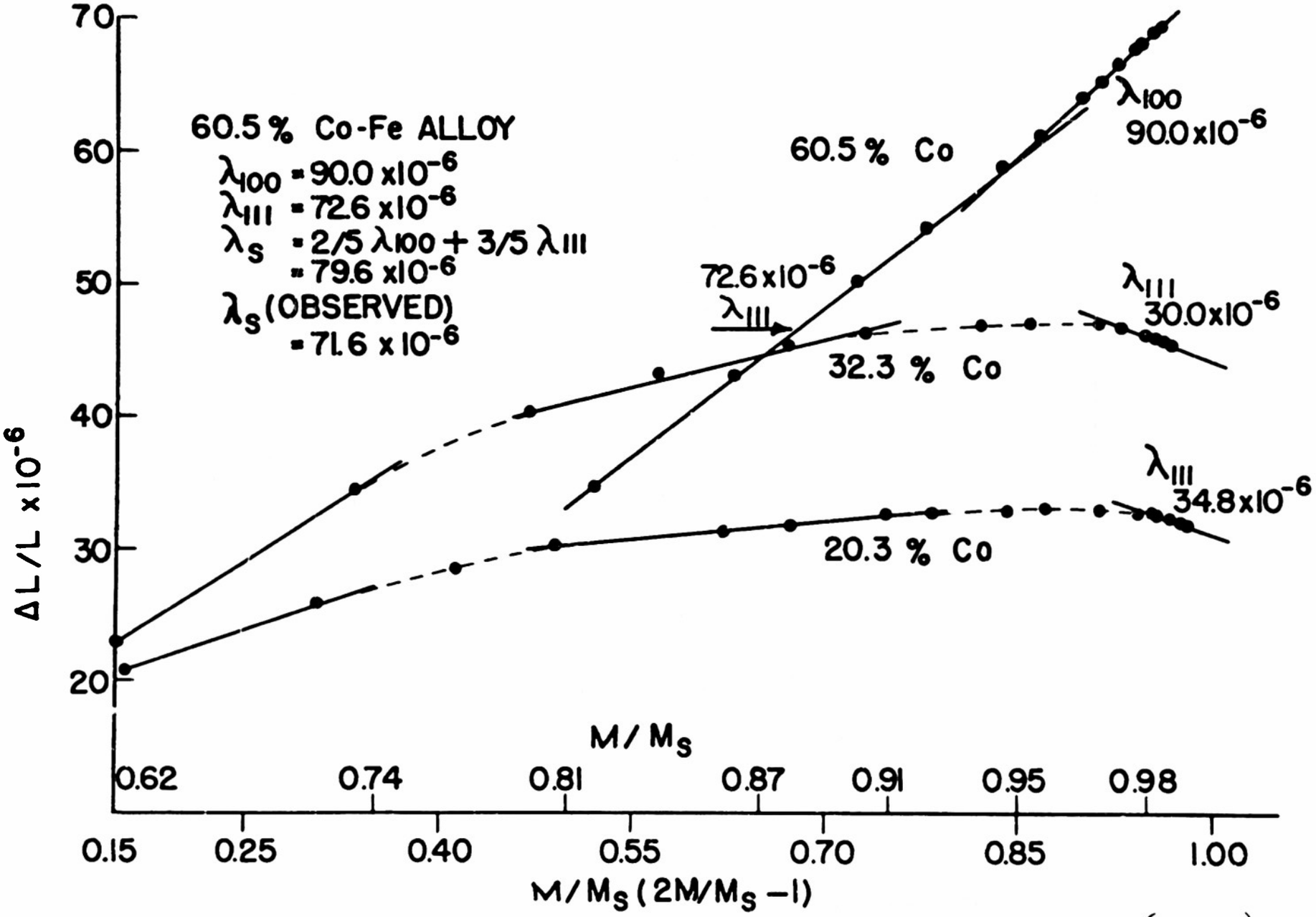

Figure 6. Magnetostriction of polycrystalline iron-cobalt alloys as a function of $\frac{M}{M_S}\left(\frac{2M}{M_S} - 1\right)$.